A Strategic Framework for the Design of Recycling Networks for Lithium-Ion Batteries from Electric Vehicles

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Abstract

In this paper, we develop a strategic framework for the design of recycling networks for spent lithium-ion batteries from electric vehicles.

The framework provides an overview of possible configuration alternatives and an integrated approach for network planning and process configuration tasks. It describes requirements on a network as well as on a process level. For that purpose, we analyse general framework conditions concerning battery return, materials, and recycling processes.

On that basis, it is possible to develop a mathematical optimisation model which enables decision support concerning the optimal evolvement of recycling sites, capacities, and processes over time.

Keywords:

Strategic Network Planning; Recycling; Spent Lithium-Ion Batteries

1 INTRODUCTION

Efficient and reliable lithium-ion batteries are a key technology for electric mobility due to their high specific power, energy density, and lifetime [1]. For the production of lithium-ion batteries, non-ferrous metals like copper, cobalt, nickel, and lithium are required, which leads to two main problems. On the one hand, the extraction or production of these metals is partly associated with high environmental impacts**.** On the other hand, the mining deposits are geographically concentrated and partially scarce. In 2008, seven countries accounted for 85 % of the world's production of mined cobalt, nearly the same holds for lithium [2]. Particularly for the EU this may lead to a shift of dependency from oil-producing countries to countries that are producing cobalt, lithium, or their alloys.

Recycling of the batteries has two main benefits. First, recycling may extend the reserves-to-production ratio of scarce resources, reduce energy-intensive primary production and therefore defer the opening or extension of mines. Natural resource savings of more than 50 % are possible [3]. Second, from a national point of view, recycling may have a stabilising, damping or even reducing effect on prices of and dependency on primary raw materials by providing a secondary feedstock. Beyond that, landfill and incineration of traction batteries are forbidden, and all batteries must be orderly recycled. For Germany, a recycling efficiency of 50 % by weight has to be achieved as from September 2011 [4; 5].

Up to now, neither recycling processes nor the necessary infrastructure exist that would allow for an industrial recycling of these batteries. Thus, a powerful recycling network must be established. Decisions about the evolvement of recycling sites, capacities, processes, and transportation links over time must be made. This is complicated due to heavy uncertainties about the development of the electric vehicle market and battery technology, as well as immature recycling processes.

Against this background, the objective of this paper is to deliver a framework for the design of a recycling network for spent lithium-ion batteries from electric vehicles. To build up the framework, in a first step we analyse specific problem characteristics. Based on this analysis, we develop a framework including the description of actors and requirements that need to be considered to design recycling networks for spent lithium-ion batteries. A planning approach will be presented. Different network constellations in dependence of identified decoupling points and two extreme market scenarios are examined. The results are discussed in a last step.

2 TERMINOLOGY AND BATTERY CONSTRUCTION

2.1 Electric Vehicles

In this contribution, the term *electric vehicles* includes any electric vehicle with a lithium-ion battery used for traction. Electric vehicles may be classified into **hybrid-electric vehicles (HEV)**, **plug-in hybrid electric vehicles (PHEV)** and **battery electric vehicles (BEV)**. The most important differences between the vehicle types are specifications like mass and materials, costs, and the lifetime of the required battery.

2.2 Lithium-ion batteries

Lithium-ion batteries are rechargeable batteries with promising properties for electric mobility, which is mainly due to their high power density and specific energy [1]. In this paper, the term *lithium-ion battery* embraces all rechargeable secondary batteries based on the exchange of lithium ions to produce electrical energy.

A lithium-ion battery used for traction is constructed on three fabrication levels: **cell, module,** and **system level**. Multiple cells are connected in series to raise voltage, resulting in modules. Modules are connected in series and in parallel to raise voltage and capacity. The resulting battery system additionally includes thermal management, monitoring, protection, charge/discharge balancing, and car integration components [1].

Lithium-ion battery cells consist of a cathode, an anode, a separator, an electrolyte, and a casing. Cathode and anode conductors are usually made of aluminium and copper foil, respectively. The cathode is coated with either a mix of lithium and other metals ($LiMeO_2$) or lithium-iron-phosphate ($LiFePO_4$). Metals (Me) used in the former are cobalt, nickel, manganese, and

J. Hesselbach and C. Herrmann (eds.), *Glocalized Solutions for Sustainability in Manufacturing: Proceedings of the 18th CIRP International Conference on Life Cycle Engineering, Technische Universität Braunschweig, Braunschweig, Germany, May 2nd - 4th, 2011*,
DOI 10.1007/978-3-642-19692-8_14,

aluminium. The anode is usually coated with graphitic or amorphous carbon, or lithium-alloying metals [6]. Anode and cathode material can be combined almost freely. All of them have both specific advantages and disadvantages related to power, lifetime, safety, and costs of the cells. Lithium-ion cells can further be classified by their type of electrolyte: liquid-type, gel-type and solid-type cells. The latter two are often referred to as lithium-polymer batteries. The type of electrolyte used determines the type of separator and vice versa.

Since hybrid electric vehicles and battery electric vehicles substantially differ in their requirements regarding power and energy, lithium-ion cells can be divided into high-power cells for HEVs and high-energy cells for BEVs. Table 1 summarises typical materials and mass portions for lithium-ion batteries. For high-energy batteries, the active materials of the anode and cathode make out the largest portion of the battery mass, followed by the electrolyte. The same holds for the costs of the materials [1].

Part	Material(s) [1; 6; 7]	Mass portions [1]	
		High-Energy	High-Power
Cell level			
Cathode conductor	Aluminium foil	2 %	5 %
Cathode active material	Lithium and cobalt, nickel, manganese, or aluminium, or a mix of them, or Lithium-Iron-Phosphate; Carbon black; Binder (PVDF)	41 %	19 %
Anode conductor	Copper foil	4 %	10 %
Anode active material	Graphitic carbon, amorphous carbon, or lithium-titanate; Binder (PVDF)	17 %	4 %
Electrolyte	Lithium salts (e. g. $LiPF_6$) dissolved in organic solvents	16 %	11 %
Separator	Polyolefins or Polyvynilidenfluorid	2 %	4 %
Cell package and others	Aluminium, stainless steel, and/or plastics	9 %	28 %
Module and system level			
Electric conductors	Copper cables	9 %	19 %
Electronics	Printed circuit assemblies including semiconductors		
Module packaging	Polypropylene		
System packaging	Aluminium, stainless steel, and/or plastics		
Total		100 %	100 %

Table 1: Typical materials for lithium-ion batteries.

3 GENERAL CONDITIONS

Figure 1 shows the generic structure of a recycling network connecting material flows between sources and sinks with different transforming processes. **Sources**, **sinks**, and **processes** of the recycling network are fraught with uncertainties. Source-related uncertainties can be classified into **quantity**, **spatial distribution**, and **specifications** of the battery returns. These parameters essentially influence decisions about total capacity, sites, and required equipment of the recycling network. Sink-related uncertainties can be classified into the three categories **product reuse**, **component reuse**, and **material reuse opportunities**. To design the network infrastructure connecting sources and sinks, the transforming processes and the resulting material flows have to be considered. Process-related uncertainties exist with respect to the optimal **combination and configuration of processes**.

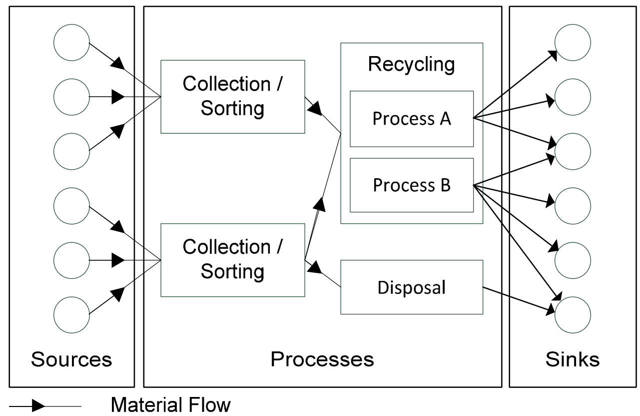

Figure 1: Generic network structure for lithium-ion battery recycling.

3.1 Uniqueness of the Planning Situation

Different recycling networks exist which could serve as a basis for a recycling network for lithium-ion batteries, including legally independent networks for vehicles, lead-acid starter batteries, portable batteries, and electronic waste.

However, these networks are not fully qualified for an efficient and environmental recycling of lithium-ion batteries. On the one hand, a high amount of spent batteries would overstrain the capacities of the mostly smallest businesses. Lithium-ion batteries are more complex, heavier, and more hazardous, hence, more difficult to treat than the mentioned products. The dangerous-good property of the batteries complicates the usage of existing transport links. On the other hand, novel and unexplored processes with high economies of scale are required which contravenes the mostly decentralised organisational form of the networks. New actors like battery material producers come into play. These circumstances would demand a restructuring of existing networks anyway.

Moreover, a particular challenge to the planning of the recycling network is the directive 2006/66/EC of the European Union. It forces battery and electric vehicle producers to conceive and establish an effective collection and recycling solution before market penetration. Because **neither the electric vehicle market nor the battery technology are evolved** to a steady state, this planning and investment situation is heavily uncertain.

3.2 Source-related uncertainties

Quantity of battery returns

Lithium-ion batteries from electric vehicles become available for recycling at the end of life of the electric vehicle or the end of life of the battery itself. Given that, the quantity of spent battery returns is particularly driven by the development of the **electric vehicle market** over time and the **battery lifetime**.

The forecast of the electric vehicle market is currently subject to many market studies. These most often use scenario technique to determine sales and stocks for specific points in time on an aggregate level [8]. For Germany, the government aims at a market penetration of 1,000,000 PHEVs and BEVs in 2020 [9]. The forecasts are significantly depending on assumptions about oil

prices, energy supply, public interest in climate change, and more stringent greenhouse gas legislation [8].

The battery lifetime is expected to be highly dependent on the used active materials and the type of vehicle the battery is used in [10]. It is estimated to be around **five years for BEVs**, **ten years for PHEVs**, and up to **fifteen years for HEVs** [10; 11]. However, these numbers are based on extrapolation, not experience, and it remains unclear how the lifetime will be distributed.

Spatial distribution of battery returns

The spatial distribution of battery returns determines transport distances and therefore costs. Decisions about sites for the collection of spent batteries and for the recycling facilities are influenced by that, given that cost minimisation or profit maximisation is preferred and all batteries must be collected.

It is not clear yet whether electric vehicles with low cruising ranges will be sold rather in urbanised than in rural areas. Based on that, not only the amount but also the spatial distribution of battery returns can be considered as highly uncertain.

Composition of battery return

Mass, size, construction, and materials of lithium-ion batteries vary dependent on their application. To give an example, a battery for a mid-sized all electric vehicles with a range of about 200 kilometres would weigh about 300 kilograms [12]. A battery for a plug-in hybrid vehicle with an electric range of 50 kilometres would weigh just about 100 kilograms [13]. This diversity and the highly uncertain technology development demands either highly flexible equipment or manual work, which tends to result in high investments or costs, respectively.

3.3 Sink-related uncertainties

Product reuse opportunities

Spent batteries from electric cars which may not be suitable for powering vehicles anymore are expected to be qualified for further stationary use, e. g. storage of energy from fluctuant power generation like wind or solar power [1]. Direct product reuse then would decelerate the return of spent batteries. However, it is not expected to happen in a large scale while the batteries are not further standardised so that they can be easily stocked, connected, and monitored outside of vehicles.

Component reuse opportunities

Standardisation is also important to enable component reuse. With standardised battery constructions, electronic interfaces, and cell types, spent batteries could be remanufactured easily by replacing worn components with new ones, resulting in as-good-as-new batteries. Additionally, cells with a similar state from different spent batteries could be assembled to lower-quality batteries, and electronic components could be used as spare parts.

Material reuse opportunities

The high cobalt-price of approximately 40 USD per kilogramme was the driver for the recycling of smaller lithium-ion batteries, but also one important reason to replace parts of the cobalt with other metals [1] or entirely move away from cobalt-containing cathode active materials. By that, the **economical incentive of recycling** may diminish. Especially lithium-iron-phosphate does not contain any cobalt or nickel (which is priced at about 20 USD per kilogramme). Lithium, in the form of lithium carbonate, is much cheaper at about 6 USD per kilogramme and the mass fraction of it on cell level is just about 3 %. From today's point of view, the costs of conditioning may excess the revenues. However, lithium prices are expected to rise as the demand is growing dramatically.

3.4 Process-related uncertainties

Recycling processes

Basically, recycling processes for lithium-ion batteries can be classified into disassembly, mechanical conditioning, hydrometallurgical conditioning, and pyrometallurgical conditioning. In **disassembly**, battery systems are broken down subsequently to module and cell level which is the prerequisite for a remanufacturing of batteries. Reusable components and large material fractions, for example the battery casing and electronics, are separated before further conditioning processes are applied to the cell materials. In **mechanical conditioning**, materials of the cells are separated by different comminution, sizing, and concentration processes. Examples are crushing, screening, and magnetic separation. **Pyrometallurgical conditioning** involves the thermal treatment of the materials at high temperatures. It can be combined with the production of steel, and of ferromanganese or other alloys. **Hydrometallurgical conditioning** typically combines leaching, solution concentration and purification, and metal recovery methods, including electrometallurgy. With hydrometallurgical processes, pure non-ferrous metals can be recovered from active materials after mechanical conditioning or from the slag of pyrometallurgical processes.

Process combinations

As listed in chapter 2.2, lithium-ion batteries can consist of a variety of different materials. Single materials can be recovered selectively with certain processes. Therefore, a **combination of different processes is necessary** to recover all relevant materials [14]. Figure 2 depicts a comprehensive overview of different process combination possibilities and the recovered materials. It includes potential decoupling points that by way of example occur if a disassembly or mechanical conditioning is intended or necessary. The prerequisite for spatial decoupling is the transportability of the intermediate products, which appears to apply mainly for cells as well as for the coating powder, and, to a less extent, for the intermediate products from pyro- and hydrometallurgical processes.

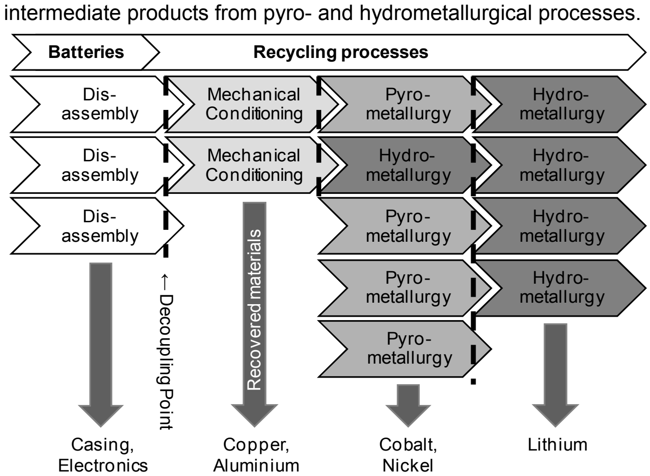

Figure 2: Combination possibilities of conditioning processes and recovered materials, based on [15].

Exemplary process combinations

A promising process to recover all valuable materials is currently subject of investigation at **LithoRec**. The analysed process begins with demounting the spent battery from the car. After transportation to a disassembly line, the battery will be disassembled to cell level. Cells will be dismantled and the volatile liquids (electrolyte with conductive salts) are absorbed for further conditioning. The coated electrodes will be separated with mechanical processes, resulting in aluminium and copper fractions and coating powder. The coating

powder, which contains the active material, will be treated in chemical conditioning processes to recover lithium, nickel, cobalt, and manganese, or lithium-iron-phosphate, which will then be available for production again. See Figure 3 for an illustration.

The LithoRec process promises to be suitable for large-scale recycling, since a large amount of materials is recovered, including battery components, all metals, and the valuable electrolyte. However, the process is still under investigation.

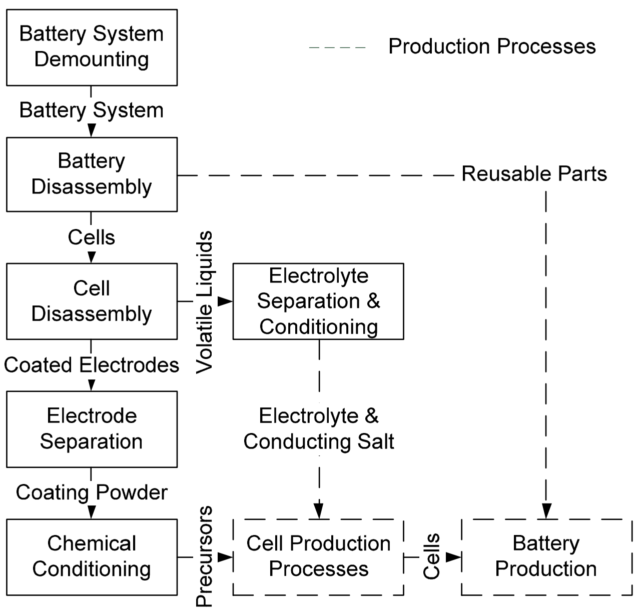

Figure 3: Combined mechanical and hydrometallurgical processes.

[3] describe a combined pyrometallurgical and hydrometallurgical recycling process for lithium-ion batteries, amongst others, which is already applied at Umicore in Sweden and Belgium. Here, a mix of different consumer lithium-ion batteries is treated in a pyrometallurgical smelting process located in Sweden, ending in a slag containing aluminium, silicium, calcium, and lithium, and an alloy of cobalt, nickel, copper, and iron. The alloy is then transported to Belgium and treated with hydrometallurgical processes to separate the materials, while the materials in the slag may be consumed in lower value production processes.

Although this process is already applied, it appears to be less suitable for recycling vehicle batteries in large scale, because valuable materials like aluminium and lithium are lost in the slag. However, in combination with subsequent hydrometallurgical processes, the recovering of these materials should be possible.

3.5 Network design and process configuration

As shown, the quantity and material specifications of the batteries which are to be used as educts fundamentally influence the eligible processes and their products (recovered materials, components, etc.). The configuration of processes, in turn, determines the spatial decoupling potential. In centralised concepts, all batteries would be treated in a single facility to realise economies of scale, resulting in low average costs due to high capacity utilisation. In contrast, a decentralised concept with many small facilities would enable short transportation links and thus low transportation costs. Processes with spatial decoupling points can provide both advantages. Decentralised pre-treatment with low economies of scale, e. g. manual disassembly, would allow for a concentration of the material flow to reduce transportation costs while reducing transport mass and volume by the separation of casings. The subsequent treatment of the cells could then be realised centrally, exploiting high economies of scale.

In summary, the processes chosen and the battery types processed determine the remanufacturing option, recoverable materials and their degree of purity as well as the spatial decoupling potential. While some combinations of processes would be able to recover new active material like $LiCoO_2$, some would result in pure cobalt and lithium-carbonate, among copper and aluminium. Thus, different sinks must be considered in each case. By this means, the **decision about processes considerably influences the network structure** and must be integrated into strategic network planning.

4 FRAMEWORK

4.1 Scope and corporative actors

The framework for the design of a network for the recycling of spent lithium-ion batteries involves different kinds of actors of the economy, namely the battery material producers, battery producers, vehicle producers, vehicle dealers, and vehicle treatment operators.

The **battery material producer** is predestined to recover the materials, since this would be near to his core competence. By this means he can influence the characterisation of the raw materials directly and, concurrently, prevent potential competitors.

Although the production of batteries may not be the core competence of vehicle producers today, it will most probably be in the future. Both, **battery producers** and **vehicle producers,** can be in charge for the correct treatment of the batteries by law, and both rely on the continuous supply with raw battery materials. So, a clear distinction is not necessary. From the producer's point of view, the recycling must be carried out in a cost-minimising or profit-maximising manner. At the same time, he is bound to collect and recycle every battery that is given to him by the end-users. Thus, he cannot minimise costs by cherry-picking, but he can share his fixed costs and use economies of scale by the joint operation of a cross-producer network.

All mentioned actors of the production chain are reliant on a continuous supply of raw materials. As mentioned in chapter 1, this may become a problem in the light of scarce and geographically concentrated resources. Given that, all producers will be interested in the supply from a secondary feedstock, even though the recovered materials may be costlier or a little less capable than primary materials.

The **vehicle dealer** is involved since he will be the first address for users of electric vehicles if their battery is spent. His task will be the replacement of the spent battery with a new one. Thus, he is interested in an easy and free-of-charge solution for the redistribution of the received spent battery, without wanting to relevantly participate in the network. Nevertheless, his participation will be needed to gather information about the battery conditions. Exactly the same holds for the **treatment operator** who is engaged with the treatment of end-of-life vehicles. He, however, will try to charge the vehicle manufacturer with additional fees for the demounting of the battery, or alternatively sell it to others.

These deliberations show that **all actors** should be interested in an optimised collection and recycling of spent batteries because the requirements of the actors mostly converge. This can only be achieved by planning and operating the network collaboratively.

For the actors, economic interests are prior. Beyond that, they have to respect legal conditions and customer requirements. Therefore, environmental aspects have to be considered as well. For this reason, this framework addresses the design of a sustainable cooperative recycling network with focus on the producers who are legally responsible for its provision.

4.2 Requirements

As shown in chapter 3, a recycling network for lithium-ion batteries is subject to many uncertain factors that change over time. This requires a long-term planning horizon, in which uncertainties must be represented adequately. Decisions with respect to the design of the network include the selection of facility locations, processes, capacities, and the point in time of their implementation. Due to the described interdependencies between optimal network structure and recycling processes, these decisions must be done simultaneously. Possible synergy effects by the inclusion of existing sites and processes of recycling networks for vehicles, starter batteries, portable batteries, and electronic waste as well as their actors have to be examined. Sites of treatment operators and vehicle dealers are sources of the networks and could be used for regional collection and even for the disassembly of spent batteries.

The processes for the recycling of the batteries, including mechanical, chemical, and thermal treatment, differ in many aspects. They must be represented in an adequately aggregated manner. Their eligibility must be assessed with respect to their differences in:

- material flows, including educts, raw materials and supplies, products, and their quality;
- energy consumption;
- environmental impacts in terms of pollutant emissions;
- investments and costs, influenced by economies of scale;
- subsequent expandability;
- degree of flexibility regarding the processable materials and volumes;
- potential spatial decoupling of single process steps;
- hazard potential, particularly for the employees.

To represent centralised and decentralised installation concepts, a multi-stage recycling network must be considered. Process-dependent material flows between sources and sinks have to be included. The structure of the network should be flexible to enable subsequent changes with respect to the combination of processes, their spatial decoupling, and capacity expansions. Furthermore, an option to store batteries to delay expenses, balance capacity utilisation, and use economies of scale has to be assessed.

4.3 Approach

The objective of our approach is to give decision support to investors regarding the provision of an optimised sustainable recycling network for lithium-ion batteries, including the selection of facility locations, processes, capacities, and the point in time of their implementation. Thus, this planning task can be classified into strategic network planning within the APS matrix.

Strategic network planning approaches have been discussed in the literature for related recycling networks with similar characteristics, for instance, vehicle recycling [16], portable battery recycling [17], electrical equipment recycling [18], and large household appliances recycling [19]. Further similarities can be found in the strategic planning of a biofuel production network presented in [20]. These approaches mainly include location, transport, and capacity decisions; the latter is explicitly regarding uncertainties and process decisions. They provide the basis for the following approach, whereas some of the requirements listed in chapter 4.2 are not satisfied. E. g. none of the approaches considers a multiple objective optimisation.

In the first step of our approach the materials which have to be recovered from the batteries due to economical, ecological, or strategic reasons will be identified. For that, the development of their prices, sources, production and mining methods, reuse potentials, and their scarcity will be analysed.

Next, known and promising conditioning processes for these materials and the possible combinations will be screened. A pre-selection is necessary to narrow down the choice. This will be done by a comparison of the processes with respect to multiple criteria, namely monetary, ecological, social, flexibility, and safety criteria. For that purpose, aggregated material and energy flow sheet models of the processes will be used. For the equipment of the preselected processes, investment needs and costs depending on different capacities will be estimated in a pre-calculation, unless they are known. Potential decoupling points of the processes will be identified to determine the possibility of a decentralised recycling network. Transportation costs regarding the batteries and the different intermediate products will be estimated. Sources and sinks of the network will be identified.

Subsequently, location factors for the collection and recycling sites will be established. Appropriate sites for recycling facilities will be classified regarding the different requirements of the various recycling processes. Again, a pre-selection of sites will be necessary. The inclusion of site-specific investments and costs will be investigated to couple interdependencies between expenses for sites, buildings, and equipment.

To give decision support, a multi-staged dynamic facility location problem with the specific requirements of reverse logistics problems will be developed and then used in a mathematical optimisation. Input data are:

- long-term battery return estimations;
- costs for transportation and storage of the batteries;
- locations of existing sources and sinks;
- distances between these locations and potential collection and recycling sites;
- costs and investment needs for facilities and processes;
- ecological factors, e. g. CO_2-emissions.

The optimisation will be done with respect to different objective functions. Prevailing uncertainties will be considered by the application of scenario technique, robust optimisation, and sensitivity analysis. The findings will end in an extensive robust investment plan for the design of a sustainable recycling network for spent lithium-ion batteries.

4.4 Exemplary Network Alternatives

Scenario A: Centralised Network

In scenario A, a centralised network with a combined pyrometallurgical and hydrometallurgical recycling process is established. Based on an unimportant electric vehicle market, the battery technology is not further developed or standardised. Because of the low amount of lithium-ion vehicle batteries produced, a shortage of resources does not occur, and cobalt-based active materials are prevalent. Only few batteries are available for recycling; consequently, a dedicated recycling network is not economical. In the network, batteries are collected from few specialised garages and brought to a centralised recycling facility that recovers materials from any kind of batteries containing cobalt or nickel to realise high capacity utilisation. Lithium, aluminium, and manganese are not recovered. The more valuable materials cobalt, nickel, and copper are recovered in high purities in a subsequent hydrometallurgical process at the same location and are sold to any kind of markets.

Scenario B: Decentralised Network

In scenario B, a decentralised network with a combined disassembly, mechanical and hydrometallurgical recycling process

is established. A prospering electric vehicle market leads to strong technology advances regarding the lithium-ion batteries. These are produced at large scale, which leads to temporary shortages of raw materials. While miscellaneous compositions are used for the production of the cells, lithium-iron-phosphate emerges as the prevailing active material. The spent batteries are collected from dealers all over the country and are brought to decentralised disassembly facilities. Here, they are first used for energy storage until depletion. Afterwards, the batteries are disassembled to cell level. All materials except for the cells are brought to regional recycling facilities. The cells are transported to a centralised recycling facility where they are subject to mechanical and hydrometallurgical recycling. New active material is produced from the recovered lithium, nickel, manganese, and cobalt, and is sold to battery producers, as well as the recovered lithium salts and the electrolyte.

5 CONCLUSION AND OUTLOOK

In this paper, we presented a strategic framework for the design of recycling networks for lithium-ion batteries from electric vehicles. We analysed prevalent uncertainties related to potential sources, sinks, and processes for the recycling. The analysis showed that in particular the quantity of the return of batteries can be considered as uncertain, due to the early development stage of the electric vehicle market and missing experience regarding the lifetime of batteries. This complicates decisions about capacities in the network. Additionally, the uncertainties in the battery material composition and the price development for recoverable materials complicate process decisions. It is not clear which processes will emerge as eligible options for the sustainable recycling of batteries. We highlighted that the decision about processes considerably influences the network structure and must be integrated into the planning of the network.

Based on that, we derived a framework, including an analysis of potential actors and requirements that must be considered designing a recycling network for lithium-ion batteries, and an integrated planning approach. Our future research will concentrate on the analysis of eligible processes and the implementation of the approach in a mathematical multi-objective optimisation model.

6 ACKNOWLEDGEMENT

The presented research work is part of the project “LithoRec – Recycling von Lithium-Ionen-Batterien”. LithoRec aims at developing and assessing sustainable processes and full life-cycle concepts for the recycling of spent lithium-ion batteries in Germany. We would like to acknowledge the support of the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety for funding the research cooperation under the reference 16EM0023.

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